



# Pathways to ultra-low platinum group metal catalyst loading in proton exchange membrane electrolyzers

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## ABSTRACT

Hydrogen is one of the world's most important chemicals, with global production of about 50 billion kg/year. Currently, hydrogen is mainly produced from fossil fuels such as natural gas and coal, producing CO<sub>2</sub>. Water electrolysis is a promising technology for fossil-free, CO<sub>2</sub>-free hydrogen production. Proton exchange membrane (PEM)-based water electrolysis also eliminates the need for caustic electrolyte, and has been proven at megawatt scale. However, a major cost driver is the electrode, specifically the cost of electrocatalysts used to improve the reaction efficiency, which are applied at high loadings (>3 mg/cm<sup>2</sup> total platinum group metal (PGM) content). Core-shell catalysts have shown improved activity for hydrogen production, enabling reduced catalyst loadings, while reactive spray deposition techniques (RSDT) have been demonstrated to enable manufacture of catalyst layers more uniformly and with higher repeatability than existing techniques. Core-shell catalysts have also been fabricated with RSDT for fuel cell electrodes with good performance. Manufacturing and materials need to go hand in hand in order to successfully fabricate electrodes with ultra-low catalyst loadings (<0.5 mg/cm<sup>2</sup> total PGM content) without significant variation in performance. This paper describes the potential for these two technologies to work together to enable low cost PEM electrolysis systems.

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## 1. Introduction

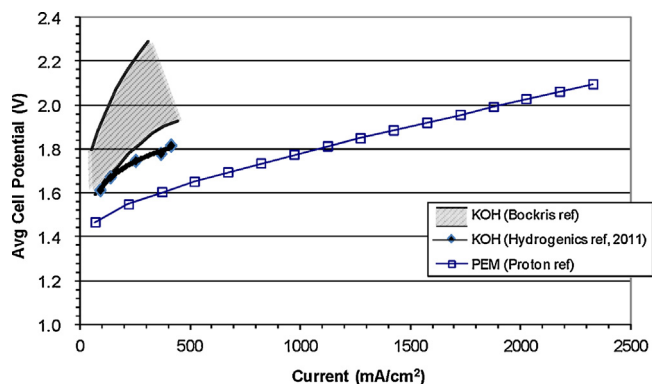
Hydrogen is one of the world's most important chemicals, with global production of about 50 billion kg/year. Currently, hydrogen is mainly produced from fossil fuels such as natural gas and coal, with CO<sub>2</sub> produced as a byproduct. Therefore, large environmental impacts are possible through development of renewable hydrogen production methods such as electrolysis of water. Energy storage

applications in Europe such as wind capture and improved bio-gas conversion efficiency are also driving significant interest in electrolysis, because penetration of renewable energy is already surpassing acceptable grid levels, and water electrolysis is the only renewable hydrogen technology mature enough to address these needs in the near term. Proton exchange membrane (PEM) electrolysis in particular is attractive due to the dynamic range, reliability, and lack of corrosive electrolyte vs. traditional liquid KOH electrolyte systems. Fig. 1 shows typical operating ranges for liquid KOH systems vs. commercial PEM systems. In both the KOH [1] and the Proton case, the polarization curves represent commercial status as of 2012–2014 and do not necessarily represent advancements demonstrated in laboratory stacks. Fig. 2 shows reliability data for Proton's cell stacks, demonstrating 60,000 h lifetimes (8 years). The older stack configuration shows a voltage decay rate which would still project to a much longer lifetime, assuming an end of life voltage of 2.6 V, while the newer design shows negligible decay even after 40,000 h of operation. Liquid KOH systems have also been claimed to demonstrate similar stack durability [2].

**Abbreviations:** BNL, Brookhaven National Laboratory; CCM, catalyst coated membrane; DOE, Department of Energy; EDS, energy dispersive spectroscopy; EDX, energy dispersive X-ray; GDE, gas diffusion electrode; GDL, gas diffusion layer; HER, hydrogen evolution reaction; HOR, hydrogen oxidation reaction; ICP-OES, inductively coupled plasma optical emission spectroscopy; OER, oxygen evolution reaction; PEM, proton exchange membrane; PGM, platinum group metal; RDE, ring disk electrode; RHE, reversible hydrogen electrode; RSDT, reactive spray deposition technology; SEM, scanning electron microscopy; TEM, transmission electron microscopy; XRD, X-ray diffraction; XPS, X-ray photoelectron spectroscopy.

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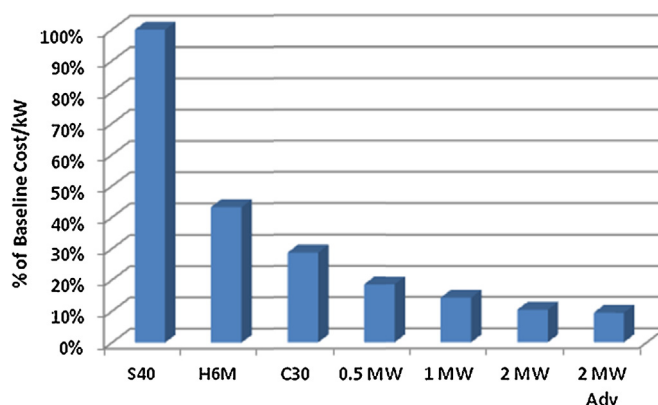


**Fig. 1.** Typical operating ranges for liquid KOH [3] and proton exchange membrane electrolyzers [4]. PEM example is at 50 °C and uses a 7-mil Nafion® N117 membrane, and 7 mg/cm<sup>2</sup> total PGM loading using platinum and IrO<sub>x</sub>.

At a small scale, the catalyst cost is not a significant cost contribution to the overall system. However, balance of plant cost per output of hydrogen decreases significantly with increases in capacity (Fig. 3). This normalized cost drops to less than 30% of the original cost with a 25-fold increase in output, and is expected to decrease a similar amount with another order of magnitude increase in power to the 2 MW system. Recently, Proton's DOE funded program to reduce the cost of the bipolar plate resulted in a significant, 40%, cell cost reduction [6].

Therefore, a major cost driver at megawatt scale becomes the catalyst coated membrane (CCM). Specifically, the cost of high loadings of platinum group metals and the current labor intensive methods of applying them to the membrane contribute over 30% of the cell stack cost (Fig. 4, left). Proton's megawatt scale product is shown in Fig. 4 (right).

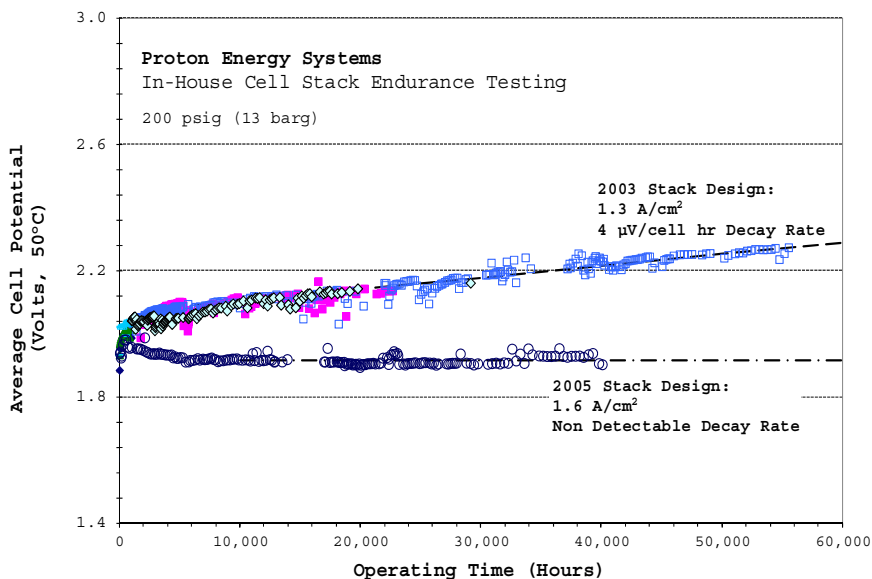
Addressing the issue of catalyst cost requires advancements in catalyst activity as well as manufacturing processes. To accomplish this goal, Proton has worked with both Brookhaven National Lab (BNL) as well as University of Connecticut. Brookhaven has developed core-shell catalysts which provide high activity at low loading in fuel cells. In collaboration between Proton and BNL, a catalyst composition suitable for the hydrogen evolution was developed. This catalyst demonstrated high performance and stability concurrent with significant reductions in catalyst loading. In parallel,



**Fig. 3.** Relative balance of plant cost per kW, normalized to 7 kW (S40) product.

Proton has been collaborating with the Maric group to develop manufacturing processes for consistent fabrication of low loaded electrodes through directly making and depositing advanced catalyst onto Nafion or gas diffusion layers (GDLs). Core-shell catalyst fabrication with RSDT or any scalable nanomanufacturing process must maintain tight control over shell thickness, down to a monolayer or two, in order to realize the shift in the electronic structure of the shell surface for enhanced catalytic activity. A separate paper focused on RSDT will provide more details on process and theory. Finally, these two advancements can work together to manufacture and deposit core-shell materials at low loadings. This paper will describe these advancements in core-shell catalysts and flame based RSDT techniques as applied separately to PEM electrolysis systems, as well as how these can work together to produce an optimized electrode.

The concept and practical application of atomic-level core-shell nanocatalysts emerged at beginning of the 21st century. By utilizing Pt spontaneous deposition on metallic Ru nanoparticles, an ultralow-Pt-content catalyst was made with 20:1 Ru:Pt atomic ratio to explore the possibility of placing all the Pt atoms at the surface of Ru particles so that they can be actively involved in hydrogen oxidation reaction [7,8]. Only 1/8 monolayer coverage of Pt was deduced based on the particle size and lattice structure observed by high-resolution transmission electron microscopy (TEM) [9]. Encouraged by the performance results, core-shell



**Fig. 2.** Durability of proton exchange membrane electrolyzers [5]. Both stacks use 10-mil Nafion® N1110 and Pt and IrO<sub>x</sub> catalysts at 10 mg/cm<sup>2</sup> total PGM loading.

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